

We claim:

1. A non-porous multi-layer membrane that is selectively permeable to hydrogen having a hydrogen source surface and a hydrogen sink surface and which comprises a hydrogen-permeable central layer and at least one catalyst layer that facilitates the dissociation of hydrogen positioned in the membrane between the central layer and the hydrogen source surface and at least one protective layer or at least one barrier layer.
2. The non-porous membrane of claim 1 wherein the hydrogen-permeable layer is a cermet wherein the metal of the cermet is a metal or alloy that is permeable to hydrogen, except that the metal or alloy is not palladium or an alloy of palladium.
3. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer is vanadium or an alloy of vanadium.
4. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer comprises: V, Nb, Ta, Zr or a mixture thereof in combination with one or more of Al, Co, Cr, Fe, Mn, Mo, Nb, Ti, Ta, Cu, Ni, Ga, Ge, Sn, Zr, Si, W, La, Be, and Hf as binary, ternary, or quaternary alloys.
5. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer is a multi-phase hydrogen-permeable material comprising at least one hydrogen-ion conducting metal oxide and at least one electron-conducting metal oxide.
6. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer is a multiphase material comprising at least one hydrogen ion-conducting phase and at least one electron-conducting phase.
7. The non-porous membrane of claim 6 wherein the electron-conducting phase is a metal phase that is not permeable to hydrogen.
8. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer

is a multi-phase hydrogen-permeable material comprising at least one hydrogen-ion conducting oxyacid salt phase and at least one electron-conducting phase

9. The non-porous membrane of claim 1 wherein the hydrogen-permeable membrane layer is a multi-phase hydrogen-permeable material comprising at least one hydrogen-ion conducting fluoride phase and at least one electron-conducting phase.

10. The non-porous membrane of claim 1 wherein the hydrogen-permeable layer further comprises a metal oxide that does not conduct hydrogen ions and does not conduct electrons.

11. The non-porous membrane of claim 1 having a barrier layer wherein the barrier layer is a non-porous hydrogen-permeable material.

12. The non-porous membrane of claim 11 wherein the barrier layer comprises a hydrogen ion-conducting metal oxide.

13. The non-porous membrane of claim 12 wherein the hydrogen-ion conducting metal oxide is a doped perovskite.

14. The non-porous membrane of claim 11 wherein the barrier layer comprises a hydrogen ion-conducting oxyacid, a hydrogen ion-conducting fluoride or a mixture thereof.

15. The non-porous membrane of claim 1 having a protective layer which is a porous metal oxide.

16. The non-porous membrane of claim 15 wherein the protective layer further contains one or more metals that dissociate hydrogen sulfide.

17. The non-porous membrane of claim 1 comprising at least two protective layers one of which comprises a high surface area metal oxide and the other of which comprises a metal oxide wherein the metal forms stable sulfides.

18. The non-porous membrane of claim 1 having a protective layer which comprises a high surface area metal oxide wherein a metal of the metal oxide forms stable sulfides.
- 5 19. The membrane of claim 1 wherein the catalyst layer is selected from the group consisting of palladium, an alloy of palladium, nickel, an alloy of nickel, a cermet comprising palladium, an alloy of palladium, nickel or an alloy of nickel and the 8B and 1B metals and mixtures and alloys thereof.
- 10 20. The membrane of claim 19 wherein the catalyst layer is selected from the group consisting of Fe, Co, Ni, Cu, Ru, Rh, Pd, Ir, and Pt.
21. The membrane of claim 1 wherein the hydrogen-permeable central layer comprises an alloy of vanadium and titanium.
- 15 22. The membrane of claim 1 wherein the hydrogen-permeable central layer comprises an alloy of vanadium and aluminum.
23. The membrane of claim 1 wherein the hydrogen-permeable central layer comprises an alloy comprising vanadium and one or more of aluminum, titanium, cobalt, molybdenum, and chromium.
- 20 24. A reactor membrane for separation of hydrogen from a hydrogen-containing feedstream which comprises any one of the membranes of claim 1.
- 25 25. A membrane reactor comprising one or more membranes of claim 1.
26. A method for separation of hydrogen from a hydrogen-containing gas which comprises the steps of:
- 30 contacting a hydrogen feedstream with the hydrogen source surface of a membrane of any of claim 1;

heating the membrane to a temperature such that hydrogen selectively permeates through the membrane to a hydrogen sink.

27. The method of claim 26 wherein the hydrogen is separated from gas mixtures containing
5 CO₂, CO, H₂S or mixtures thereof.

28. The method of claim 26 wherein the hydrogen is separated from gasified coal, water-gas-shift mixtures, reformed petroleum products, or reformed methane, butane, ethanol or ammonia.

10 29. A method for carrying out oxidation-reduction reactions which comprises the steps of:
contacting a hydrogen-containing species with the hydrogen source side of a membrane
of claim 1;

contacting the hydrogen sink side of the membrane with a species to be reduced, or a sweep gas or vacuum to remove hydrogen; and

15 heating the membrane to an operational temperature suitable for the oxidation-reduction reaction to proceed such that hydrogen is permeated through the membrane and such that the hydrogen-containing species is oxidized.

30. The method of claim 29 wherein the membrane is heated to an operational temperature
20 ranging from about 250 to about 800°C.

31. The method of claim 29 wherein the oxidation-reduction reaction is a hydrocarbon dehydrogenation reaction, an aromatic coupling reaction, an oxidative dimerization or oligomerization reaction, or hydrogen sulfide decomposition.

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